Cadmium and lead in vegetable and fruit produce selected from specific regional areas of the UK

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Abstract

Cadmium and lead were determined in fruit and vegetable produce (~1,300 samples) collected from a field and market basket study of locally grown produce from the South-West of Britain (Devon and Cornwall). These were compared with similarly locally grown produce from the North-East of Britain (Aberdeenshire). The concentrations of cadmium and lead in the market basket produce were compared to the maximum levels (ML) set by the European Union (EU). For cadmium 0.2% of the samples exceeded the ML, and 0.6% of the samples exceeded the ML for lead. The location of cadmium and lead in potatoes was performed using laser ablation ICP-MS. All tested samples exhibited higher lead concentrations, and most exhibited increased concentrations of cadmium in the potato skin compared to the flesh. The concentrations of cadmium and lead found in fruits and vegetables sampled during this study do not increase concern about risk to human health.

Keywords: cadmium; lead; fruits; vegetables; maximum levels.

Capsule: Cd and Pb was determined in fruit and vegetable produce, from the SW and NE of Britain, it was determined that less than 0.6% of the produce exceeded guides values for these 2 elements.

Introduction

Cadmium and lead can both have adverse effects on human health. Cadmium is a chronic potent nephrotoxin as well as a class one carcinogen, and is associated with a range of other severe diseases (EFSA, 2012; Clemens et al., 2013). Acute and chronic lead exposures have been linked to a number of conditions (WHO, 2010; Fewtrell et al., 2003). Inorganic lead has also been classified as a compound which is probably carcinogenic to humans (IRAC, 2006). One of the main routes of cadmium intake for humans is through the consumption of food containing cadmium (Kabata-Pendias and Mukherjee, 2007; Clemens et al., 2013).

Toxic metals, including cadmium and lead, can be introduced to soils from various sources (Nicholson et al., 2003). These include sewage sludge, livestock manures, inorganic fertilisers, natural geogenic anomalies, base and precious mining activity, and industrial “wastes” (Nicholson et al., 2003). The main route of lead and cadmium input to agricultural land in England and Wales is through atmospheric deposition, and for cadmium the second highest route is through the application of phosphate fertilisers (Nicholson et al., 2003).

The European Union (EU) has set maximum levels (MLs) of both cadmium and lead in fruit and vegetables based on produce class, all based on wet weight (European Commission Regulation (EC) No. 1881/2006 (as amended)). For cadmium the standard for leafy vegetables, fresh herbs, celeriac, and all cultivated fungi is 200 ng g^{-1}. For stem vegetables, root vegetables, and potatoes it is 100 ng g^{-1}, while for vegetables and fruits excluding leafy vegetables, and other products (listed before), it is 50 ng g^{-1}. The MLs set by the EU for lead in berries and small fruits is 200 ng g^{-1} and 100 ng g^{-1} for other fruits. The ML for lead in brassicas and leaf vegetables is 300 ng g^{-1} while in other vegetables (including peeled potatoes) it is 100 ng g^{-1}. Previously, Weeks et al. (2007) conducted a survey of 12 metals (including cadmium and lead) from 12 allotment sites in the UK (6 urban and 6 rural), measuring a total of 251 fruit and vegetable samples. The study identified no produce above the ML for either cadmium or lead. The highest concentration of lead was found in a blackcurrant sample (160 ng g^{-1}), and the highest concentration of cadmium was in a spinach sample (40 ng g^{-1}). Yuan et al. (2014) conducted a health risk assessment of cadmium from food in China. They determined that the concentration of cadmium in vegetables ranged from below the limit of detection (LOD) to 150 ng g^{-1}, with a mean concentration of 13.3 ng g^{-1}, while the mean concentration of cadmium in fruit was 0.8 ng g^{-1}, and ranged from below LOD to 9.1 ng g^{-1}. In the study they concluded that mean cadmium concentrations of the food groups were below the Chinese maximum limits, but some individual samples did exceed the maximum limits, and the main dietary sources of cadmium were meat, rice, vegetables, and flour (Yuan et al., 2014). In a study looking at the health risk caused by the ingestion of cadmium and lead from a range of vegetables, where cadmium varied from 20 - 670 ng g^{-1} and lead ranged from 120 – 6540 ng g^{-1}, it was determined that the ingestion of the vegetables was unlikely to be a risk to the target population (Garg et al., 2014).

The aim of this current survey was to evaluate the concentrations of cadmium and lead in produce grown in the South-West (SW) of Britain (Devon and Cornwall) and the North-East (NE) of Britain (Aberdeenshire). The SW of Britain was picked as it has an extensive history of mining, while the NE of Britain has very limited, and localised, metal extraction industries. Growing produce on mine impacted soils can have an effect on the accumulation of heavy metals in the edible parts of the plants, leading to a hazardous accumulation of arsenic and lead (Gonzalez-Fernandez et al., 2011). Here, for both the SW and NE of Britain, basket surveys of local produce were performed, and in the SW a field survey was also conducted to link soil concentrations of cadmium and lead with concentrations of these elements in the produce. Form the data the following were tested; are the concentrations of cadmium and lead higher in vegetables and fruits grown in an area with historical mining? Is the concentration of cadmium and lead in the soil a predictor of concentrations of cadmium and lead in vegetables and fruits? Do peeled vegetables and fruits have lower concentrations than unpeeled vegetables and fruits? Additionally, laser ablation (LA)-ICP-MS experiments were done to identify the location of the elements within the produce.

Materials and Methods

Sample collection

Locally produced fruit and vegetables in retail outlets from the SW of Britian and NE Britian, as well as field crops and soil from the SW, were sampled as described in Norton et al. (2013). The basket survey selected locally grown produce from a wide range of retailers as described in Norton et al. (2013). Samples were prepared by washing the produce to a level normally used in food preparation, in a kitchen sink using local tap water. Once dry the samples were diced in a food processor then frozen before further processing. For items eaten with and without skin (potatoes, root vegetables, apples, etc.), duplicate samples were collected and one was peeled and the other unpeeled, to reflect different dietary exposures.

The samples were prepared as commonly consumed, *i.e.* unpeeled for apples and courgettes, and peeled for potatoes, swedes, parsnip, carrots, beetroots, and squashes. A duplicate sample was also prepared with the other preparation method (*i.e.* peeled apples, unpeeled potatoes, etc.), referred to as the “alternative preparation”. The SW Britain basket survey consisted of 630 samples with 207 alternative preparations, while the NE Britain basket survey consisted of 190 samples with 69 alternative preparations.

To establish the link between cadmium and lead in the produce and the concentration of cadmium and lead in the soil, both produce (fruit and vegetables in season) and soil in farmers’ fields in the SW Britain geographic areas were sampled, with the farmers’ permissions, as described in Norton et al. (2013). A total of 174 soil samples were analysed along with corresponding crops, as well as 56 alternative preparations for produce eaten either peeled or unpeeled.

To determine the total cadmium and lead in the skin and flesh of baked potatoes 20 potato samples were collected. For each potato sample three individual potatoes were baked and samples prepared as described in Norton et al. (2013). To compare the concentrations of cadmium and lead in the flesh and the skin a paired t-test was performed on the matching skin and flesh.

Cadmium and lead analysis

Samples were pureed in a blender, which was thoroughly washed between each sample. Pureed produce samples were accurately weighed into 50 mL polyethylene centrifuge tubes and oven dried at 70 ^{o}C, and the moisture content determined. The weight of wet material was to give an estimated dry weight (accurately weighed) of between 0.2 – 0.3 g. For digestion 2.5 mL of concentrated nitric acid was added to each sample and then incubated overnight. Trace reagent analysis grade reagents were used throughout. Prior to microwave digestion 2.5 mL of hydrogen peroxide was added to each tube and then the samples were digested using a Microwave Accelerated Reaction System (MARS, CEM); digestions were performed as described in Norton et al. (2013). Four certified reference materials (CRM) were used: 1x IC-INCT-MPH-2 mixed Polish herbs, 1x NIST-1568a rice flour, 1x CTA-OTL-1 Oriental tobacco leaves, and 1x NCS ZC73012 cabbage.

Sieved soil samples were weighed (0.1 g) into glass digest tubes and 2.5 mL of concentrated nitric acid was added to each tube. The samples were left overnight with the acid. Hydrogen peroxide (2.5 mL) was added to each tube and the digest tubes were then transferred to a digest-heating block set at 100 ^{o}C; after 1 h the temperature was increased to 120 ^{o}C, then after 1 h the temperature was increased to 140 ^{o}C. The samples were then digested for 4 h. The samples were transferred to 15 mL centrifuge tubes and made up to 10 mL, followed by a 1:10 dilution of the digest. For each soil batch one reaction blank, one spike, and one soil CRM (NCS ZC73007) were used.

Cadmium and lead analysis was performed by ICP-MS (Agilent Technologies 7500). Rhodium (10 µg L^{-1}) was run on an external line as the internal standard. The cadmium and lead standards ranged from 0.1 – 300 µg L^{-1}.

The mean concentrations and recoveries for a number of different CRMs for cadmium and lead are presented in Table 1. For the cabbage (NCS ZC73012), mixed Polish herbs (IC-INCT-MPH-2), and oriental tobacco (CTA-OTL-1) CRMs 35 independent digestions were performed, and for the rice flour (NIST-1568a) CRM 28 independent digestions were performed. No value is given for lead in the rice flour CRM as the value is not certified. The results from the CRMs were reliable and showed good consistency between digestion batches.

Laser Ablation – Inductively Coupled Plasma – Mass Spectroscopy

Potatoes from both the SW and NE survey were collected for determining the localisation of cadmium and lead within whole potatoes. After sampling, all produce was stored fresh at 5 ^{o}C for up to one week before analysis. Whole produce samples were washed with tap water. Samples were then diced (~1 cm^3) and three skin containing diced samples were randomly chosen per item. The cubes were then flash frozen using liquid nitrogen and stored frozen at -20 ^{o}C until sectioning. Sections were prepared from each batch at -15 ^{o}C using a cryostat (Model OTF including microtome 5030, Bright). A random cube from each batch was mounted on a sample holder and fixed using Tissue-Tek O.C.T. paste (Sakura Finetek Europe). A microtome was used to prepare thin-sections of up to 35 µm depth. The slices were thaw-mounted onto microscope slides and allowed to air-dry at room temperature (for 1 – 2 hours depending on the thickness of the slice), and subsequently stored frozen at -20 ^{o}C until analysis. Each sample was analysed by LA-ICP-MS (New Wave model UP-213), interfaced with ICP-MS (Agilent Technologies, model 7500c), and masses (m/z) carbon [13], sulphur [34], silicon [29], cadmium [112], and lead [208] were monitored. Silicon was monitored in order to delineate the slide from the sample. Carbon and sulphur were used to verify that the thickness of the ablation was relatively constant. Three ablation lines were performed for each section and the data averaged for plotting. The ablation always started at least 200 µm outside the sample, on the skin side. As the structure of each sample (density and porosity) will determine how much of it is ablated, LA-ICP-MS cannot be used for quantitative analysis.

Results

Market Basket Survey

The concentrations of cadmium and lead for the market basket produce collected from SW Britain are presented in Figure 1 and Tables 2 and 3. Produce is grouped into fruit and vegetable groups, and only produce with three or greater independently collected samples is presented. The concentration of cadmium in the SW produce ranged greatly with concentrations below the LOD to 123 ng g^{-1}. For cadmium in produce the MLs set by the EU are determined by fruit and vegetable class (European Commission Regulation (EC) No. 1881/2006). The standard for cadmium in leafy vegetables, fresh herbs, celeriac, and all cultivated fungi is 200 ng g^{-1}. For stem vegetables, root vegetables, and potatoes it is 100 ng g^{-1} cadmium, and for vegetables and fruits excluding leafy vegetables, and other products (listed before) it is 50 ng g^{-1} cadmium. For the leafy vegetables no values exceeded the cadmium ML of 200 ng g^{-1}, with the highest value for this class of vegetables being 123 ng g^{-1} (chard). The highest stem vegetable had a cadmium concentration of 58 ng g^{-1} (leek), below the ML of 100 ng g^{-1}, and none of the root vegetables exceeded the ML either; the highest was a beetroot (56.9 ng g^{-1}). For the fruit category none exceeded the maximum of 50 ng g^{-1} cadmium, and for the other vegetables none exceeded 50 ng g^{-1} cadmium.

The concentration of lead in produce from the SW of England ranged from below the LOD to 529.4 ng g^{-1}. The ML set by the EU for lead in fruit and vegetables is determined by vegetable and fruit class (European Commission Regulation (EC) No. 1881/2006 (as amended)). The ML for lead in berries and small fruits is 200 ng g^{-1} and 100 ng g^{-1} for other fruits. The ML for lead in vegetables (including peeled potatoes) is 100 ng g^{-1}. For the berries and small fruits two strawberry samples exceeded the 200 ng g^{-1} lead ML (Table 3). For the lead ML of 300 ng g^{-1} for brassicas and leaf vegetables no sample exceeded this value. For the lead ML of 100 ng g^{-1} for the other produce types one sample exceeded this limit (beetroot).

The cadmium and lead concentrations for the market basket produce for the classes of produce collected from the NE of Scotland is presented in Figure 2 and Tables 4 and 5. For the leafy vegetables a single spinach sample (479 ng g^{-1}) exceeded the ML of 200 ng g^{-1} cadmium (data not presented in Figure 2 and Table 4 as less than 3 spinach samples were collected in the NE survey). None of the stem vegetables exceeded the cadmium ML of 100 ng g^{-1} and only a single root vegetable (beetroot, 101 ng g^{-1}) exceeded the limit. For the fruit category none exceeded the maximum of 50 ng g^{-1} cadmium, and for the other vegetables none exceeded 50 ng g^{-1} cadmium. For the berries and small fruits no samples exceeded the 200 ng g^{-1} lead ML and no sample exceeded the 300 ng g^{-1} lead ML for brassicas and leaf vegetables. For the 100 ng g^{-1} lead ML for the other produce types two samples (these were a beetroot and a carrot) exceeded this limit.

For nine different classes of fruit and vegetables a comparison was conducted, between fruit and vegetable, unpeeled and peeled, for lead (Figure 3) and cadmium (Figure 4). Statistical analysis was performed using a paired t-test for each produce category to take into account the variation in concentration of lead and cadmium observed at different locations. For cadmium and lead apples (n=17), beetroots (n=14), carrots (n=32), courgettes (n=22), cucumbers (n=3), parsnips (n=9), and squashes (n=7) showed no significant difference between peeled and unpeeled. For potatoes (n=79) and swedes (n=23) there was significantly more cadmium and lead in the vegetables that had not been peeled.

Relationship between produce and soil cadmium and lead

Soil cadmium concentrations ranged from 0.13 - 2.79 µg g^{-1} with a mean of 0.59 µg g^{-1}, and soil lead concentration ranged from 9.2 – 916 µg g^{-1} with a mean of 78.2 µg g^{-1}, for the field samples collected in the SW of Britain. Correlations between produce cadmium concentration and soil cadmium concentration (Supplementary figure 1) identified a single significant positive correlation between cadmium in cabbage and soil cadmium ($P<0.001$, $r= 0.658$, $n= 25$). No correlation between soil cadmium and soft fruit cadmium content could be performed as all the soft fruit cadmium concentrations were below the LOD. For lead concentration in produce only the root vegetables (both unpeeled ($P<0.001$, $r = 0.743$, $n= 19$) and peeled ($P<0.001$, $r = 0.93$, $n= 19$)) correlated with soil lead concentration (Supplementary figure 2). No correlation between soil lead and soft fruit lead content could be performed as all the soft fruit lead concentrations were below the LOD.

Comparison of baked potato skin and flesh

From the SW England survey, 20 potato samples were selected for the determination of cadmium and lead in the skin and flesh of baked potatoes. There was significantly ($P<0.001$) more cadmium and lead in the skins of the baked potatoes compared to the flesh (Supplementary figure 3). On average the concentration of cadmium in the potato skins was 23.7 ng g^{-1} compared to 8.3 ng g^{-1} in the flesh (f.wt). On average the concentration of lead in the potato skins was 162.7 ng g^{-1}, while in the flesh all values were below the LOD of 8 ng g^{-1} (f.wt).

Localisation of cadmium and lead in root vegetables/tubers

The localisation of cadmium and lead in potatoes was performed using LA-ICP-MS. A cross-section was taken across the potato from the skin following a trajectory towards the centre of the produce sample. All potatoes sampled exhibited higher lead concentrations in the skin than in the flesh (Figure 5). Most samples contained higher cadmium concentrations in the skin than in the flesh (Figure 5). The peaks for both these elements cover a depth of approximately 400 to 900 µm inside the potatoes.

Discussion

In general, the produce from the SW basket survey had equivalent concentrations of cadmium and lead compared to the produce from the NE basket survey, and in both cases there were a limited number of samples that were above the EU ML (European Commission Regulation (EC) No. 1881/2006, as amended). For cadmium the produce with the highest concentrations are loosely classed into open leaf structure vegetables, *i.e.* those produce which have a large surface area in relation to mass. Out of all the produce analysed (as normally prepared) only two samples (0.2% of the total samples) exceeded the ML (European Commission Regulation (EC) No. 1881/2006, as amended) for any of the produce classes (both from the NE survey). For lead a total of 5 samples (0.6% of the total samples) exceeded the ML for their class of produce across the two basket surveys. A previous fruit and vegetable survey in the UK reported that of the 251 produce samples they collected none exceeded the ML for cadmium and lead; the highest cadmium value was in a spinach sample (40 ng g^{-1}) and for lead in a blackcurrant sample (160 ng g^{-1}) (Weeks et al., 2007). Although not investigated as part of this study, more washing of the produce may help in removing some metal contamination. In the current study produce was washed using tap water to a level normally used in food preparation. However, some produce, due to their comparatively large surface area to mass (e.g. those classified as open leaf structure vegetables), are susceptible to elemental contamination arising from soil/dust deposition, which may increase concentrations of the elements (Norton et al., 2013). In a study by Ferri et al. (2015) it was observed that unwashed spinach leaves had a higher concentration of heavy metals compared to washed spinach leaves. This has also been observed in a number of other studies where thorough washing decreases the concentration of heavy metals in leaves (Caselles, 1997; Oliva and Valdés, 2006). The issue of atmospheric deposition could also in some part explain the lack of correlations between the soil heavy metal concentration and plant concentration, as soil/dust deposition may have a large influence on the final measured concentration.

When comparing the different preparation methods only potatoes and swedes had lower cadmium and lead concentrations in the peeled produce compared to the unpeeled produce. For potatoes this is further confirmed by the potato baking experiment where the concentrations in the baked potato skin were greater than in the potato flesh (Supplementary Figure 3). The location of the cadmium and lead was further explored by LA-ICP-MS in the potatoes and it was identified that there is an increase in the concentration of cadmium and lead within the first 1 mm of the potatoes (Figure 5). The findings that the cadmium concentration is greater in the non-peeled *vs.* peeled potatoes, the concentration of cadmium is greater in the skin of baked potatoes compared to the flesh of the potatoes, and that the cadmium is located within 400 - 900 µm of the outside of the potatoes is in agreement with Corguinha et al. (2013) who found that the cadmium concentration was higher in the potato peel rather than the tuber. The data also indicates that the peel of the potatoes has a far greater concentration of lead compared to the flesh of the potato as revealed by the comparison of peeled *vs.* non-peeled potatoes (Figure 3), LA-ICP-MS (Figure 5), and the concentration of lead in the skin and flesh of baked potatoes (Supplementary figure 3). These results indicate that a proportion of the contaminating metals could be removed by peeling the potatoes to at least a depth of 1 mm.

The range observed in concentrations of cadmium and lead in the individual species could be down to differences in both the cultivars and the field sites on which they were grown. In a study on cadmium accumulation in water spinach it was observed that there was a greater than two fold difference in shoot cadmium for 32 cultivars grown on the same soil under the same conditions (Xiao et al., 2015). Corguinha et al. (2013) observed that for potatoes there were significant differences in cadmium concentrations for different cultivars, and that the same cultivar grown at different locations accumulated different concentrations of cadmium. These variation in cultivars and locations may partly explain the lack of correlation between soil cadmium concentrations and produce cadmium concentrations (Supplementary figure 1), as different cultivars were sampled across different fields.

The lack of observed correlations between soil and plant heavy metal concentration has also been observed in a number of other studies (Menzies et al., 2007; Ferri et al., 2015). The absence of correlations between soil cadmium and plant cadmium concentration and soil lead and plant lead concentration may be due to a number of factors already discussed, *i.e.* cultivar variation and atmospheric deposition. However, there are a large number of other potential reasons why there is a lack of a relationship between soil and plant heavy metal concentration. As this survey was done across a wide range of different locations it is expected that the soil properties, which affect the availability and uptake of metals from soils, will be different at different sites. Soil pH has been identified as an important factor in cadmium accumulation in plants (Adams et al., 2004; Zheng et al., 2011; Zhao et al., 2010; Ding et al., 2013 Liu et al., 2015). For example, Liu et al. (2015) demonstrated that wheat varieties accumulated more cadmium in acidic soils compared to alkaline soils. Other important soil factors include organic carbon content, cation exchange capacity, and clay content, as well as the oxide content of Fe, Al, and Mn (Zheng et al., 2011; Zhao et al., 2010; Ding et al., 2013;). As these factors would vary across the different sampling locations, this may be a reason for the lack of correlations between soil and plant heavy metal concentration.

There are many potential sources of cadmium and lead to agricultural land (Nicholson et al., 2003). Apart from atmospheric deposition the largest contribution of cadmium to English and Welsh agricultural soils is inorganic fertilisers, specifically phosphate fertilisers (Nicholson et al., 2003).The long term application of these fertilisers could potentially have led to an accumulation of heavy metals in the soil (Cheraghi et al. 2012). It has been observed that the long term use of phosphate fertiliser led to a moderate accumulation of some heavy metals (including lead) and a significant accumulation of cadmium in the soils of Hamadan province in Iran (Cheraghi et al. 2013). Cheraghi et al. (2013) identified that potatoes grown on heavy metal contaminated soils were contaminated with heavy metals, however, there was not a health risk in eating produce from these soils. An additional factor that should be considered is that the concentrations of cadmium and lead should not be taken in isolation, as previously it has been highlighted that the region in which the South-West of Britain survey was conducted had vegetables and fruits with increased concentrations of arsenic (Norton et al., 2013). The small number of samples that exceed the ML indicate a need to continue monitoring the concentration of cadmium and lead (and other potentially harmful heavy metals) in produce, but do not indicate a systemic issue with the concentrations of these elements in produce.

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Table 1. Certified reference material recoveries for total cadmium and lead analysis

Element	CRM	Certified value (ng g^{-1})	Determined value (mean ± s.e.m; ng g^{-1})	Recovery (%)
Cd	Cabbage (NCS ZC73012)[1]	35±6	33±01	93.5
	Mixed Polish herbs (IC-INCT-MPH-2) [1]	199±23	176±2	88.6
	Oriental tobacco (CTA-OTL-1) [1]	1120±120	1031±12	92.1
	Rice flour (NIST-1568a)[2]	22±2	18±1	83.5
	Soil (NCS ZC 73007)[3]	250±20	340±50	135.2
Pb	Cabbage (NCS ZC73012)[1]	190±30	173±19	91.1
	Mixed Polish herbs (IC-INCT-MPH-2) [1]	2160±230	1596±48	73.9
	Oriental tobacco (CTA-OTL-1) [1]	4910±800	3444±70	70.1
	Rice flour (NIST-1568a)[2]	-	NA	-
	Soil (NCS ZC 73007)[3]	61000±2000	44336±1677	72.7

[1] Total number of independent digests = 35; [2] Total number of independent digests = 28; [3] Total number of independent digests = 12

Table 2. Average cadmium concentrations in produce collected for the SW basket survey.

Category	Produce	n	mean cadmium (ng g^{-1} f.wt)	min cadmium (ng g^{-1} f.wt)	median cadmium (ng g^{-1} f.wt)	max cadmium (ng g^{-1} f.wt)
Compact florets and leaves	Broccoli	20	6.4	1.8	5.1	22.8
	Brussels sprouts	5	4.5	1.8	4.7	6.2
	Cabbage	66	3.8	1.8	1.8	21.2
	Cauliflower	28	3.4	1.8	1.8	9.9
	Romanesque	7	4.1	1.8	1.8	10.3
Open leaf structure	Chard	4	52.9	26.3	30.9	123.4
	Greens	18	9.4	1.8	9.0	40.4
	Kale	9	10.1	1.8	6.5	39.2
	Lettuce	42	16.6	4.0	12.2	49.3
	Salad, mix	6	30.6	13.7	24.1	61.5
	Spinach	8	47.7	11.0	44.5	112.6
Legumes	Broad beans	17	1.8	1.8	1.8	1.8
	Peas	3	1.8	1.8	1.8	1.8
	Edible pods	8	1.8	1.8	1.8	1.8
Roots and tubers	Beetroot	18	18.1	1.8	14.7	56.9
	Carrot	36	14.1	1.8	15.3	33.9
	Parsnip	12	10.1	1.8	7.1	32.4
	Potato	82	7.2	1.8	6.5	21.5
	Swede	26	4.5	1.8	4.1	9.4
Squashes	Courgette	23	1.8	1.8	1.8	1.8
	Cucumber	4	1.8	1.8	1.8	1.8
	Squash	8	3.3	1.8	3.1	5.5
Various	Asparagus	9	1.8	1.8	1.8	1.8
	Leek	31	13.9	1.8	10.7	58.1
	Onion	6	12.6	1.8	8.5	29.6
	Peppers	3	3.8	1.8	4.6	5.0
	Rhubarb	9	18.0	1.8	12.9	39.9
	Tomato	12	3.6	1.8	2.8	10.5
Fruit	Apples	17	1.8	1.8	1.8	1.8
	Currants	4	1.8	1.8	1.8	1.8
	Gooseberries	12	1.8	1.8	1.8	1.8
	Raspberries	11	1.8	1.8	1.8	1.8
	Strawberries	47	2.4	1.8	1.8	17.2

Table 3. Average lead concentrations in produce collected for the SW basket survey.

Category	Produce	n	mean lead (ng g^{-1} f.wt)	min lead (ng g^{-1} f.wt)	median lead (ng g^{-1} f.wt)	max lead (ng g^{-1} f.wt)
Compact florets and leaves	Broccoli	20	4.3	4.0	4.0	10.8
	Brussels sprouts	5	4.0	4.0	4.0	4.0
	Cabbage	66	8.7	4.0	4.0	297.6
	Cauliflower	28	4.4	4.0	4.0	10.8
	Romanesque	7	5.7	4.0	4.0	10.9
Open leaf structure	Chard	4	59.2	4.0	48.6	135.7
	Greens	18	16.6	4.0	10.2	76.5
	Kale	9	11.9	4.0	12.8	29.3
	Lettuce	42	27.0	4.0	13.1	233.5
	Salad, mix	6	115.5	20.5	94.6	236.1
	Spinach	8	79.8	4.0	28.2	282.2
Legumes	Broad beans	17	4.0	4.0	4.0	4.0
	Peas	3	4.0	4.0	4.0	4.0
	Edible pods	8	4.7	4.0	4.0	9.8
Roots and tubers	Beetroot	18	33.7	4.0	9.7	354.8
	Carrot	36	23.3	4.0	15.1	72.1
	Parsnip	12	18.7	4.0	11.3	59.5
	Potato	82	5.0	4.0	4.0	26.8
	Swede	26	4.0	4.0	4.0	4.0
Squashes	Courgette	23	5.2	4.0	4.0	16.7
	Cucumber	4	4.0	4.0	4.0	4.0
	Squash	8	4.0	4.0	4.0	4.0
Various	Asparagus	9	5.2	4.0	4.0	14.8
	Leek	31	5.5	4.0	4.0	27.3
	Onion	6	4.0	4.0	4.0	4.0
	Peppers	3	6.2	4.0	4.0	10.7
	Rhubarb	9	53.7	11.0	36.0	133.1
	Tomato	12	4.0	4.0	4.0	4.0
Fruit	Apples	17	23.3	4.0	4.0	322.3
	Currants	4	5.3	4.0	4.0	9.2
	Gooseberries	12	25.0	4.0	4.0	191.8
	Raspberries	11	4.7	4.0	4.0	12.2
	Strawberries	47	27.2	4.0	4.0	529.4

Table 4. Average cadmium concentrations in produce collected for the NE basket survey.

Category	Produce	n	mean cadmium (ng g^{-1} f.wt)	min cadmium (ng g^{-1} f.wt)	median cadmium (ng g^{-1} f.wt)	max cadmium (ng g^{-1} f.wt)
Compact florets and leaves	Brussels sprouts	4	6.8	1.8	6.8	11.8
	Cabbage	14	3.2	1.8	1.8	10.0
	Cauliflower	5	1.8	1.8	1.8	1.8
Open leaf structure	Kale	4	14.7	8.9	12.2	25.6
	Salad, mix	4	99.6	35.0	89.9	183.8
Legumes	Edible pods	9	1.8	1.8	1.8	1.8
Roots and tubers	Beetroot	11	35.4	6.4	29.1	101.0
	Carrot	12	26.6	6.4	18.8	77.4
	Potato	28	18.5	1.8	16.9	45.5
	Swede	7	9.6	4.7	9.8	13.5
	Turnip	5	14.6	6.1	6.8	26.9
Squashes	Courgette	5	2.4	1.8	1.8	4.6
Various	Leek	11	26.3	1.8	18.4	83.6
	Onion	10	6.2	1.8	4.5	16.5
	Rhubarb	4	7.3	1.8	7.1	13.2
	Tomato	7	2.5	1.8	1.8	6.4
Fruit	Apple	6	1.8	1.8	1.8	1.8
	Currents	5	5.0	1.8	6.0	9.3
	Gooseberries	5	2.2	1.8	1.8	3.8
	Raspberries	5	4.3	1.8	5.0	6.4
	Strawberries	7	2.9	1.8	1.8	6.2

Table 5. Average lead concentrations in produce collected for the NE basket survey.

Category	Produce	n	mean lead (ng g^{-1} f.wt)	min lead (ng g^{-1} f.wt)	median lead (ng g^{-1} f.wt)	max lead (ng g^{-1} f.wt)
Compact florets and leaves	Brussels sprouts	4	36.9	4.0	4.0	135.8
	Cabbage	14	5.4	4.0	4.0	14.7
	Cauliflower	5	4.0	4.0	4.0	4.0
Open leaf structure	Kale	4	118.8	18.7	102.0	252.4
	Salad, mix	4	30.8	11.0	25.5	61.3
Legumes	Edible pods	9	5.3	4.0	4.0	11.3
Roots and tubers	Beetroot	11	30.2	4.0	15.5	141.8
	Carrot	12	24.1	4.0	14.3	116.6
	Potato	28	7.6	4.0	4.0	67.7
	Swede	7	5.9	4.0	4.0	10.9
	Turnip	5	7.6	4.0	4.0	17.4
Squashes	Courgette	5	11.9	4.0	4.0	29.8
Various	Leek	11	13.8	4.0	4.0	96.7
	Onion	10	5.1	4.0	4.0	14.7
	Rhubarb	4	33.7	14.4	23.9	72.5
	Tomato	7	4.0	4.0	4.0	4.0
Fruit	Apple	6	14.2	4.0	4.0	61.0
	Currents	5	14.7	4.0	9.7	36.7
	Gooseberries	5	4.9	4.0	4.0	8.4
	Raspberries	5	4.0	4.0	4.0	4.0
	Strawberries	7	5.3	4.0	4.0	13.0

Figure 1. Cadmium concentrations (a) and lead concentrations (b) in produce basket survey conducted in the SW of Britain.

Figure 2. Cadmium concentrations (a) and lead concentrations (b) in produce basket survey conducted in the NE of Britain.

Figure 3. Percentage of lead detected in the unpeeled produce compared to the peeled produce. Significant was determined using a paired t-test (* = $P < 0.05$). Error bar is the standard error of the mean.

Figure 4. Percentage of cadmium detected in the unpeeled produce compared to the peeled produce. Significant was determined using a paired t-test (* = $P < 0.05$). Error bar is the standard error of the mean.

Figure 5. LA-ICP-MS traces for potato.

Supplementary Figure 1. Relationship between produce cadmium concentration and soil cadmium. (A) unpeeled potatoes, (B) peeled potatoes, (C) unpeeled root vegetables, (D) peeled root vegetables (E) cabbage, (F) cauliflower and broccoli, (G) open structure produce.

Supplementary Figure 2. Relationship between produce lead concentration and soil lead. (A) unpeeled potatoes, (B) peeled potatoes, (C) unpeeled root vegetables, (D) peeled root vegetables (E) cabbage, (F) cauliflower and broccoli, (G) open structure produce.

Supplementary Figure 3. Cadmium (a) and lead (b) concentration in the flesh and skin of baked potatoes.